Temperature dependence of polarization relaxation in semiconductor quantum dots

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The decay time of the linear polarization degree of the luminescence in strongly confined semiconductor quantum dots with asymmetrical shape is calculated in the frame of second-order quasielastic interaction between quantum dot charge carriers and LO phonons. The phonon bottleneck does not prevent significantly the relaxation processes and the calculated decay times can be of the order of a few tens picoseconds at temperature $T \simeq 100$K, consistent with recent experiments by Paillard et al. [Phys. Rev. Lett. **86**, 1634 (2001)].

PACS numbers: 78.67.Hc, 72.25.Rb, 63.20.Ls

The discrete nature of the energy spectrum in semiconductor quantum dots (QDs) is supposed to lead to a strong suppression of spin relaxation[1] where promising applications for new spin dependent electronic devices have been predicted[2,3]. The current interest in manipulating semiconductor spins for applications is based on the ability to control and maintain spin coherence over practical length and time scales. Optical pumping experiments have indeed given good indications of a slowing down of the carrier spin relaxation processes in QD compared to bulk or quantum well structures.[4,5] Recently, a detailed time-resolved investigation of the intrinsic spin dynamics in InAs/GaAs QDs under strictly resonant excitation has been reported by Paillard et al.[6] They demonstrated that at low temperature the carrier spins are totally frozen on the scale of the exciton lifetime. A rapid temporal decay of the linear polarization degree is, however, found above 30K.

As a possible intrinsic mechanism for temporal decay of the linear polarization, we propose here the second-order quasielastic interaction between QD carriers and LO phonons as sketched in Fig. 1(b). A similar mechanism was suggested by Uskov et al.[7] to explain the temperature dependence of a broadening of the zero-phonon line. We will show that such relaxation processes are not suppressed (strongly) by the "phonon bottleneck" and can lead to a decay time of the linear polarization of some tens of picoseconds at $\simeq 100$K.

Let us first recall that self-organized QDs are usually strained and have an asymmetrical shape with a height smaller than the base size. The upper valence band in such QDs with the zinc–blende lattice is split into a heavy-hole band with the angular momentum projections $j_{h,z} = \pm 3/2$ and a light-hole band with $j_{h,z} = \pm 1/2$ at the center of the Brillouin–zone (here the growth direction z is chosen as the quantization axis). The conduction band is s–like, with two spin states $s_{e,z} = \pm 1/2$. The heavy-hole exciton quartet is characterized by the total angular momentum projections $J_z = \pm 1, \pm 2$. The radiative states $J_z = \pm 1$ and nonradiative ones, $J_z = \pm 2$, are split by the electron-hole exchange interaction (so–called singlet–triplet splitting Δ_{st}).[8–10] In the case of an asymmetric confinement potential in the plane of QDs the symmetry of the system is lowered and the exchange interaction is no longer isotropic.[1,11] As a result, both doublets are split into singlets, as is shown schematically in Fig. 1. The radiative doublet $|\pm 1\rangle$ is split by an anisotropic exchange into the states labeled $|X\rangle = (|1\rangle + \imath|-1\rangle)/\sqrt{2}$ and $|Y\rangle = (|1\rangle - \imath|-1\rangle)/\sqrt{2}$, giving rise to optical transitions which are linearly polarized along the [110] and [1$\bar{1}$0] directions, respectively.[12,13] Continuous wave single dot spectroscopy experiments have clearly evidenced these two linearly polarized lines in self-organized InGaAs QDs.[13] An analogous splitting of the exciton states has been observed in studies of single QD's formed at GaAs/AlGaAs interfaces.[14] The observation of the "optical orientation-optical alignment" or "alignment-orientation" transformation in a magnetic field makes it possible to determine the magnitude of the splitting without resolving the fine–structure spectrally.[6,12,15] The anisotropic exchange splitting originates from the elongation of the QDs.[1,12,16] The calculated and measured magnitudes of this splitting reach some tens or even hundreds of μeV.[6,12–14]

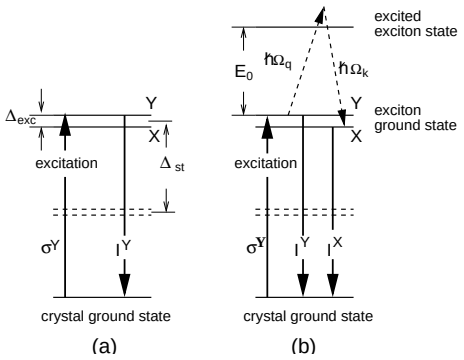

FIG. 1. Schematic diagram showing (a) the sublevels of the exciton ground state and (b) the second-order phonon scattering process within the radiative doublet via the first excited state. Optically-inactive states are shown by dashed lines.

In the case of (pulsed) resonant excitation which is linearly polarized, say, along the Y–direction (σ^Y- excitation), an exciton in the Y–sublevel is created. If there is no relaxation to the X–sublevel, the following emission occurs from the same Y- sublevel, as is shown in Fig. 1(a), and the linear polarization degree of the luminescence $P_{lin} = (I^Y - I^X)/(I^X + I^Y)$ (where I^X and I^Y denote the X and Y linearly polarized luminescence components) remains constant within the exciton lifetime, $P_{lin} = 1$. The possible relaxation process is shown schematically in Fig. 1(b). This is the second-order exciton-phonon scattering process in which one phonon is absorbed and another one is emitted. The scattering events occur via the excited states of the exciton (with negligible exchange splitting) which can couple to both X– and Y–exciton states. These processes lead to the exciton transitions between the ground Y– and X– sublevels and result in the temporal dependence of P_{lin}. The (complete) rate equations governing the populations f_Y and f_X of the Y- and X-sublevels read

$$\frac{df_Y}{dt} = -\frac{f_Y}{\tau_{rad}} - \frac{f_Y}{\tau_{sc}} + \frac{f_X}{\tau_{sc}} ,$$
$$\frac{df_X}{dt} = -\frac{f_X}{\tau_{rad}} - \frac{f_X}{\tau_{sc}} + \frac{f_Y}{\tau_{sc}}, \qquad (1)$$

where τ_{sc} is the scattering time for an exciton relaxing from the $Y(X)$–state to the $X(Y)$–state and τ_{rad} is the radiative recombination time. For the relevant initial conditions $f_Y(0) = f_0$ and $f_X(0) = 0$, Eqs. (1) lead to an exponential decay of the polarization degree $P_{lin} = (f_Y - f_X)/(f_Y + f_X) = \exp(-t/\tau_{pol})$ with $\tau_{pol} = \tau_{sc}/2$.

As was noted above, in experiments by Paillard et al.[6] no decay of P_{lin} is observed on the exciton lifetime scale at $T < 30$K, but the decay time of P_{lin} drops from ~ 3.5ns at 40K down to ~ 50 ps at 80K. In order to explain these findings we consider the contribution of LO phonons to the second-order scattering processes above. In this case a strong (nonlinear) temperature dependence of a relaxation time τ_{sc} can be expected in the temperature range k_BT smaller than the LO phonon energy $\hbar\Omega_0$.

Our calculation of the polarization decay time is based on the Fermi golden rule for second order processes. To simplify matters, we restrict ourselves to a certain range of parameters where the following conditions hold:
(i) We assume that the phonon energy $\hbar\Omega_0$ is in the vicinity of the excitation energy E_0 from the exciton ground state doublet (labelled "0") to the first excited state (labelled "1") so that contributions of higher excited states can be neglected in the first approximation. As a result, we have

$$\frac{1}{\tau_{sc}} = \frac{2\pi}{\hbar} \sum_{\vec{q},\vec{k}} \Big| \frac{M^{01}_{\vec{q}}\, M^{10}_{\vec{k}}}{E_0 + \hbar\Omega_{\vec{k}} + \imath\Gamma/2} + \frac{M^{01}_{\vec{k}}\, M^{10}_{\vec{q}}}{E_0 - \hbar\Omega_{\vec{q}} + \imath\Gamma/2} \Big|^2 \times N_{\vec{q}}\,(N_{\vec{k}} + 1)\, \delta(\Delta_{exc} - \hbar\Omega_{\vec{k}} + \hbar\Omega_{\vec{q}}), \quad (2)$$

where $\vec{k}, \Omega_{\vec{k}}, \Gamma$ and $N_{\vec{k}} = 1/(e^{\hbar\Omega_{\vec{k}}/k_BT} - 1)$ are the phonon wave vector, frequency, linewidth, and thermal distribution function, respectively.
(ii) To evaluate the exciton–phonon matrix elements $M^{01}_{\vec{q}}$, we restrict ourselves to the Fröhlich interaction[17] of bulk LO phonons with carriers in strongly confined flat QDs (strained in the growth direction). For a strong confinement, the exciton states are reasonably well approximated by noninteracting electron–hole pair states. In addition, the electron mass is much smaller than the heavy–hole mass and the first excited exciton state is mainly composed of a ground state electron and an excited hole. As a result, the electron does not couple to phonons because of the orthogonality of the initial and final hole states. Thus, $M^{01}_{\vec{q}}$ reduces to the matrix element constructed with the hole envelope wave functions $\Psi^h_0(\vec{r})$ and $\Psi^h_1(\vec{r})$,

$$M^{01}_{\vec{q}} = \imath\, \frac{(2\pi e^2\hbar\Omega_0)^{1/2}}{q\sqrt{V\kappa}}\, \langle \Psi^h_0(\vec{r}) | e^{\imath\, \vec{q}\, \vec{r}} | \Psi^h_1(\vec{r}) \rangle, \qquad (3)$$

where $q = |\vec{q}|$, V is the normalization volume, and $1/\kappa = 1/\varepsilon_\infty - 1/\varepsilon_0$ (ε_0 and ε_∞ denote the static and high–frequency limits of the dielectric function).
(iii) Equation (3) is mostly affected by the oscillatory behavior of the exponential function rather than by the detailed shape of the wave functions. Therefore, we may furthermore neglect a weak lateral anisotropy of the QD and, for simplicity, consider a parabolic potential of the form $\sim \alpha_\parallel^4(x^2 + y^2) + \alpha_z^4 z^2$. The potential strengths $\alpha_\parallel$ and α_z are inversely proportional to the base length $L_\parallel$ and a height L_z of the QD, respectively. In this case the hole envelope wave functions are given by

$$\Psi^h_0(\vec{r}) = \frac{\alpha_\parallel \alpha_z^{1/2}}{\pi^{3/4}}\, \exp\Big\{-\frac{\alpha_\parallel^2}{2}(x^2 + y^2) - \frac{\alpha_z^2}{2} z^2\Big\},$$
$$\Psi^h_1(\vec{r}) = \sqrt{2}\alpha_\parallel\, x\, \Psi^h_0(\vec{r}). \qquad (4)$$

In addition, we have neglected the heavy–hole–light–hole mixing because in the case of strained flat QDs the energy separation between the first heavy–hole and light–hole levels is large compared to the lateral confinement energies.
(iv) It follows from Eqs. (3,4) that only LO–phonons with small wave numbers $q \lesssim \alpha_\parallel \sim L_\parallel^{-1}$ contribute substantially to the scattering rate Eq. (2). Therefore, the phonon dispersion can be ignored in the Bose functions, $N_{\vec{k}}, N_{\vec{q}} \to N_0$. In the delta–function, however, the dispersion is essential and it will be approximated by an isotropic relation, $\hbar\Omega_{\vec{k}} = \hbar\Omega_0 - vk^2$. Below, we restrict our consideration to energy window $0.5\hbar\Omega_0 \geq |E_0 - \hbar\Omega_0| \geq 0.05\hbar\Omega_0 \approx 1.5$meV ($\hbar\Omega_0 \simeq 30$meV). In this case $|E_0 - \hbar\Omega_0|$ is larger than the exchange splitting Δ_{exc}, the effective width of the phonon band $v\alpha_\parallel^2$, as well as the phonon line width Γ.[18] Therefore, these contributions can be neglected in the energy denominators of Eq. (2).

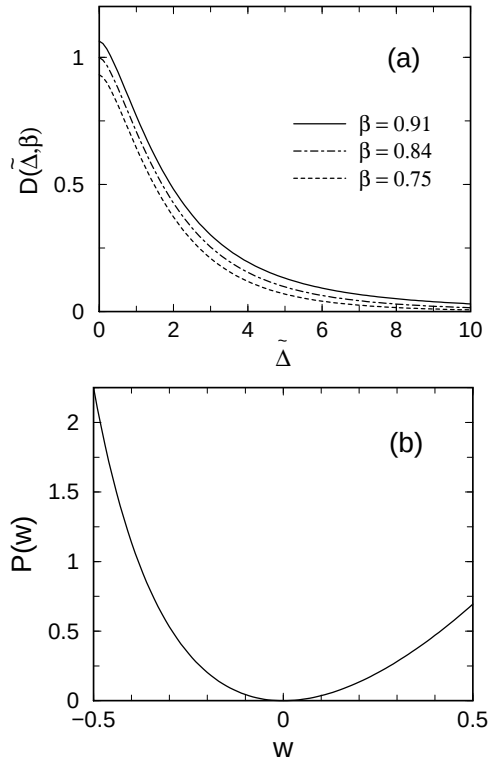

FIG. 2. Phonon (a) and level detuning (b) functions.

After some algebra, we obtain our main result

$$\tau_{pol} = \tau_0 \, \frac{1}{N_0(N_0+1)} \, \frac{P(w)}{D(\tilde{\Delta},\beta)}, \tag{5}$$

$$\tau_0 = \sqrt{2}(2)^6 \pi \frac{\kappa^2 \hbar v}{e^4}, \tag{6}$$

$$P(w) = \Big[\frac{w(w+2)}{(w+1)}\Big]^2, \tag{7}$$

$$D(\tilde{\Delta},\beta) = e^{-\tilde{\Delta}} \int_0^\infty dx x^2 e^{-x^2} F(\frac{x^2\beta}{2}) \times F(\beta(\tilde{\Delta}+\frac{x^2}{2}))\sqrt{\tilde{\Delta}+\frac{x^2}{2}}, \tag{8}$$

$$F(z) = \sqrt{\pi/z}\,\mathrm{erfi}(\sqrt{z})\,(1+1/2z) - e^{\sqrt{z}}/2\sqrt{z}, \tag{9}$$

where τ_0 is the characteristic decay time of the QD which depends on the electron–phonon coupling strength κ and the phonon dispersion parameter v. The parameter $w = (E_0 - \hbar\Omega_0)/\hbar\Omega_0$ defines the "phonon detuning" between the interlevel spacing in the QD and the LO phonon energy, whereas $\tilde{\Delta} = \Delta_{exc}/(2v\alpha_\parallel^2)$ defines the "mismatch" between the radiative level separation and the effective width of the phonon band[19,20]. The parameter $\beta = 1-(L_z/L_\parallel)^2$ measures the anisotropy of the QD (for flat QDs under consideration $\beta > 0$ and close to one) and $\mathrm{erfi}(z) = \mathrm{erf}(\imath z)/\imath$ is the imaginary error function[21].

From the structure of Eq. (5) we can draw qualitative conclusions about the influence of temperature, level and phonon–detunings, and size and shape of the QD on the polarization decay time.

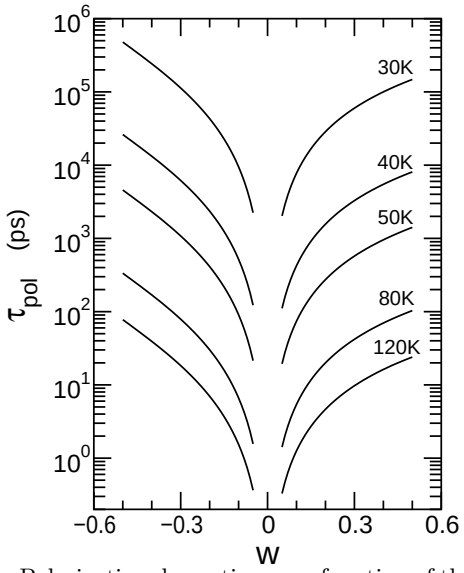

FIG. 3. Polarization decay time as a function of the phonon detuning parameter for $\tilde{\Delta} = 5$ and $\beta = 0.75$.

The temperature dependence is determined by the phonon number N_0 and τ_{pol} decreases exponentially with temperature if $k_B T \ll \hbar\Omega_0$.

The level detuning function for flat QDs with the anisotropy parameter $\beta = 0.91(L_z/L_\parallel = 0.3)$, $\beta = 0.84(L_z/L_\parallel = 0.4)$, and $\beta = 0.75(L_z/L_\parallel = 0.5)$ is shown in Fig. 2(a). $D(\tilde{\Delta},\beta)$ decreases monotonically with $\tilde{\Delta}$ and has an exponential limit for $\tilde{\Delta} \gg 1$. Note that $D(\tilde{\Delta},\beta)$ enters in Eq. (5) due to the condition of the energy conservation during the scattering process (cfm. δ-function in Eq. (2)). Furthermore, the level splitting Δ_{exc} plays the role of the energy difference $(\hbar\Omega_0 - E_0)$ when compared with an inelastic first-order scattering process.[22] As it is well known, such processes are strongly suppressed by the phonon bottleneck effect. The second–order processes under consideration are, however, more efficient. The point is that an anisotropic exchange splitting $\Delta_{exc} \sim 0.1$meV is by itself relatively small[6,13], while some specific conditions are required to obtain similar magnitudes for the energy difference $(\hbar\Omega_0 - E_0)$. In addition, the parameter $\tilde{\Delta} = (\Delta_{exc}/v\alpha_\parallel^2)$ is not expected to change very strongly with the lateral size of the QD due to the size dependence of the exchange splitting Δ_{exc}.[12,23] Thus, $\tilde{\Delta}$ is determined mostly by the lateral anisotropy of the QDs and can be of the order of unity (using for Δ_{exc} and $v\alpha_\parallel^2$ the values $\Delta_{exc} \sim 0.1$meV and $v\alpha_\parallel^2 \sim 0.01$meV, noted above, one obtains that $\tilde{\Delta} \sim 5$). It can be seen in Fig. 2(a) also that the level detuning function $D(\tilde{\Delta},\beta)$ is modified by changing the ratio $L_z/L_\parallel$. For instance, it increases by a factor of ~ 1.3 at $\beta \sim 0.91$ compared to $\beta \sim 0.75$ in the case of $\tilde{\Delta} = 2$, by a factor of ~ 1.9 at $\tilde{\Delta} = 5$, and by a factor of ~ 2.6 at $\tilde{\Delta} = 7$.

The size dependence of polarization relaxation is determined by the phonon detuning function $P(w)$ which is shown in Fig. 2(b). The nonexponential dependence of $P(w)$ on w results in a relatively weak dependence of the polarization decay on the phonon detuning. In addition, even at relatively large detunings, e.g., $w \simeq 0.5$ or $w \simeq -0.3$, $P(w)$ is of the order of unity. The relaxation occurs for both positive ($E_0 > \hbar\Omega_0$) and negative ($E_0 < \hbar\Omega_0$) detunings but there is some asymmetry.

Below we give some numerical estimates of the polarization decay time in InAs QDs which can be considered as typical for III–V compounds. Material parameters used are: $\hbar\Omega_0 \simeq 30.2$meV, $\epsilon_0 \simeq 15.15$, $\epsilon_\infty \simeq 12.25$,[24] and $v \simeq 0.1(\text{nm})^2\text{meV}$[19] which implies $\tau_0 \simeq 0.07$ps. For $T \ll 350$K the number of phonons N_0 is very small so that τ_{pol} decreases exponentially with temperature, e.g., by a factor of $\simeq 80$ between $T = 40$K and $T = 80$K which is in agreement with experiment.[6] Because the numerical values of phonon and level detuning, as well as the QD anisotropy, are usually not known exactly from experiments, w, $\tilde{\Delta}$ and β are used as adjustable parameters. As an illustration, the decay time of the polarization degree is shown in Fig. 3 as a function of the phonon detuning parameter w at different temperatures. The decay time τ_{pol} drops from a few hundred (units) of nanoseconds at $T = 30$K down to a few tens (and even units) of picoseconds at $T = 120$K, depending on the phonon detuning. For $w = -0.15$ (i.e. $E_0 - \hbar\Omega_0 \simeq -4.5$meV), $\tilde{\Delta} = 7$ and $\beta = 0.75$ which seem to be plausible for the experiments by Paillard et al.,[6] the calculated decay times $\tau_{pol} \simeq 3.5$ns (40K) and $\tau_{pol} \simeq 44$ps (80K) closely agree with the experimental results.

In conclusion, the second-order quasielastic interaction between charge carriers and LO phonons in strongly confined asymmetric QDs is identified as an intrinsic mechanism of the temporal decay of the linear polarization degree of the luminescence. Despite of the apparent obstruction by the phonon bottleneck and the level detuning effects, the proposed mechanism leads to decay times of the order of a few tens of picoseconds (or even smaller) at temperature $T \simeq 100$K which are in agreement with experiments. The relaxation processes are more efficient in flat QDs with a weak lateral anisotropy. Equation (5) may be useful for the optimation of QD–structures for application in spintronic devices.

This work was supported by the Center for Functional Nanostructures (CFN) of the Deutsche Forschungsgemeinschaft (DFG) within project A2.

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[19] We estimate the LO–phonon dispersion constant v as $v \simeq \Delta_{ph}(a/\pi)^2$, where Δ_{ph} is a width of the phonon band and a is the lattice constant. For typical values of $\Delta_{ph} \simeq 5$meV and $a \simeq 0.5$nm we have $v \sim 0.1\text{meV nm}^2$. The lateral size of a typical QD is usually more than an order of magnitude larger than a, so that the effective width of the phonon band is typically $v\alpha_\parallel^2 \sim 0.01$meV.

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